

# The Primordial Radionuclides Activity Concentrations and Associated Minerals in Rocks from Selected Quarries in Northern Uganda

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### Abstract

The primordial radionuclides (PR) activity concentrations and associated minerals in 20 rock samples from Gulu and Amuru district quarries were determined using Na (Tl) gamma ray spectrometer and Transmitted Light Microscope (TLM). These were carried out at Physics and Geology Departments of Makerere University, Kampala in Uganda. The PR activity concentrations, the gamma dose rates, and the radiological hazard indices were all determined to assess the radiological hazards and risks of the exposure on the quarry workers and on the inhabitants. The minerals were obtained to determine whether minerals containing PR were highly radioactive. The computed PR activity concentrations from the five quarries ranged from  $4.1 \pm 1.7$  to  $207.6 \pm 6.5$  for  $^{238}$ U,  $7.0 \pm$ 2.0 to 452.0  $\pm$  2.3 for <sup>232</sup>Th, and 8.6  $\pm$  2.0 to 2055.4  $\pm$  73.1 Bq kg<sup>-1</sup> for <sup>40</sup>K, respectively with averages of 57.1  $\pm$ 4.7, 122.3  $\pm$  11.1, and 914.2  $\pm$  30.3 Bq kg<sup>-1</sup>. These values were found to be above the world limit of 33.0, 45.0 and 420.0 Bq kg<sup>-1</sup> by 1.7, 2.7, and 2.2 factors. The computed average values of absorbed gamma dose rates (D), annual effective dose equivalent outdoor ( $\underline{E}_{o}$ ), excess lifetime cancer risk (*ELCR*), radium equivalent activity  $(\underline{Ra}_{eq})$ , external hazard index  $(H_{ex})$ , internal hazard index  $(H_{in})$  and gamma index (I) for the five quarries were  $134.58 \pm 10.13$  nGy h<sup>-1</sup>,  $0.34 \pm 0.03$  mSv yr<sup>-1</sup>,  $1.07 \pm 0.09$  x  $10^{-3}$ ,  $294.08 \pm 17.88$  Bq kg<sup>-1</sup>,  $0.80 \pm 0.06$ ,  $0.93 \pm 0.06$ , 0.06, 0.06, 0.06, 0.06, 0.06, 0.06, 0.06, 0.060.05 and 1.35  $\pm$  0.09, respectively. The computed average values of D,  $E_0$  and ELCR for the five quarries were above the world limit by factors of 2.2, 4.9, and 3.7, respectively. Therefore, quarry workers and the inhabitants were exposed to PR hence significant health risks associated with induced cancer and gene mutation. However, average values of  $Ra_{eq}$ ,  $H_{ex}$ , and  $H_{in}$  were found to be below the world limit except for one of the quarry sites.

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Although the studied rocks could be used as building materials without causing external diseases and respiratory infections, the average value of, *I*, showed that studied rocks should be used in moderation to limit the radiological hazards except for Palukere rocks. Mineralogical analyses has also shown the presence of quartz, potassium feldspar, plagioclase, biotite, muscovite, and opaque minerals. Quartz was the major mineral while the rest were accessories. The results of this study could be used by different institutions such as Uganda Atomic Energy Council (AEC) that protects the environment from dangers resulting from ionizing radiation. This body could assist to advise quarry workers on good practices. The National Environmental Management Authority (NEMA) that controls environmental systems should ensure that Gulu and Amuru districts work in line with set principles on environment, and where necessary to enforce the quarry by-laws and ordinances. To urge the Directorate of Geological Survey and Mines (DGSM) to also conduct geological studies for valuable radioactive minerals in the study areas of Gulu and Amuru districts. The Uganda Ministry of Health (MoH) experts could also carry out additional studies to establish the correlation of cancer infections and quarry works. This would inform the government on corrective measures that protects the population of the region.

Keywords: Primordial Radionuclides; Risk Parameters; Average Values; Quarry Sites; GDM 20 series.

#### 1. Introduction

The common radiation processes occur naturally from minerals (terrestrial source). But also, a small fraction comes from man-made elements such as nuclear weapon testing, accidental release of Chernobyl, and medical and industrial misuse [1]. Natural sources of radiation account for 85% of annual exposure dose to the world population while man-made sources account for only 15% [2]. The natural radionuclides from terrestrial source are referred to as primordial radionuclides (PR). These radionuclides are as old as the age of the earth. The most significant of these are the radionuclides of <sup>238</sup>U and <sup>232</sup>Th radioactive series and <sup>40</sup>K [3]. Minerals containing <sup>238</sup>U and <sup>232</sup>Th radioactive series were reported to be highly radioactive. Many studies carried out directly linked significant amount of PR to rocks [4]. The concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K depended on the types of rock from which the soils originated. Higher radiation levels were always found in igneous rocks such as granite and lower levels in sedimentary rocks. There were some exceptions, however, as some shales and phosphate rocks had a relatively high content of radionuclides [1, 4, 5]. Exposure to PR in rocks comes from three main routes: (i) internal exposure due to the inhalation of inert gas (<sup>222</sup>Rn) and the short-lived products of PR. (ii) the external irradiation with gamma rays from <sup>238</sup>U, <sup>232</sup>Th and their progenies and <sup>40</sup>K. (iii) the inhalation and ingestion of ore dust from quarries [5,6]. Much as the concentrations of PR were widely distributed in the environment, the levels depended on the local geological conditions, and as a result varied from one region to another [4, 5, 6, 7]. Since PR are unevenly distributed, the knowledge of their distribution in rocks help in radiation protection and measurement [4]. Several studies reported that the processing of rocks into required aggregates release low dose radionuclides into the environment [3, 15, 16]. Although exposure to low dose radiations had little effects on human, however, being exposed to for a longer period was reported to induce cancer, gene mutation and in some cases, eventual death to the victims [15]. Studies on environmental radioactivity, as result, gained significant interest worldwide as reported below.

A study by [10] detected the activity concentrations of <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K in the granitic rock samples and

estimated the radiological dangers associated with these rocks. The obtained data on activity concentrations for  $^{238}$ U (610 ± 1730 Bq kg<sup>-1</sup>),  $^{232}$ Th (110 ± 69 Bq kg<sup>-1</sup>) and  $^{40}$ K (1157 ± 467 Bq kg<sup>-1</sup>) in the granitic samples were higher than the recommended worldwide averages of 33.0, 45.0 and 420 Bq kg<sup>-1</sup> set by [17]. The radioactive levels found in the samples were caused by altered radioactive materials trapped inside granite faults. The exposure to gamma radiation from the rocks were assessed via various radiological parameters, such as radium equivalent content (856 Bq kg<sup>-1</sup>), absorbed dose rate (D) in the air (396 nGy h<sup>-1</sup>), and annual effective dose for either outdoor (0.48 mSv  $y^{-1}$ ) or indoor (1.9 mSv  $y^{-1}$ ). Based on the analysis, the radioactive levels in the examined granitic surpassed the acceptable limits; therefore, they were not safe to use in building and infrastructure applications and may cause adverse health effects. In one study [4], activity concentrations ranged from 251.72 to 1096.2 Bq kg<sup>-1</sup> for <sup>232</sup>Th, from 494 to 2593.5 Bq kg<sup>-1</sup> for <sup>226</sup>Ra, and from 1314.6 to 1846.7 Bq kg<sup>-1</sup> for <sup>40</sup>K. The obtained radiological data showed that the average internal and external hazard indices were 9.11 and 5.78, respectively, which highly exceeded the permissible limits of unity. The samples collected from these areas were not safe and could not be used as a construction material because it possessed a great radiological threat to population and great care when handling was required. In a study conducted by [14] on natural radioactivity in soil and water from Likuyu Village in the neighborhood of Mkuju Uranium deposit, radioactivity levels of 30 soil samples and 20 water samples were determined using low level gamma spectrometry. The average radioactivity concentrations obtained in soil samples for <sup>238</sup>U (51.7 Bq kg<sup>-1</sup>), <sup>232</sup>Th (36.4 Bq kg<sup>-1</sup>), and <sup>40</sup>K (564.3 Bq kg<sup>-1</sup>) <sup>1</sup>) were higher than the worldwide average concentrations value of these radionuclides reported by [17]. The average activity concentration values of <sup>238</sup>U (2.35 Bq L<sup>-1</sup>) and <sup>232</sup>Th (1.85 Bq L<sup>-1</sup>) in water samples were\_similar and comparable to their mean concentrations in the control sample collected from Nduluma River in Arusha. A study by [16] in Kilembe copper - cobalt mines and some selected residences around Kampala-Uganda using high purity germanium (HPGe) spectrometer, showed that activity concentrations of Uranium and Thorium ranged from 50 to 300 Bq kg<sup>-1</sup> and from 5 to 50 Bq kg<sup>-1</sup>, respectively. The reported average value of <sup>238</sup>U was 187.0 Bq kg<sup>-1</sup>, well above the world average of 33.0 Bq kg<sup>-1</sup> set by [17]. Mining and processing of mineral ores was found to have enhanced the levels of natural radionuclides in the mine resulting in higher background gamma radiation levels in and around the mines. Similar results were also reported by [3]. Also, [15] studied the radioactivity levels and dose rates due to natural radionuclides in rocks from selected mining areas and quarries in Eastern Uganda using a NaI gamma detector. The report showed that the specific activities of <sup>238</sup>U, <sup>232</sup>Th and  $^{40}$ K ranged from 13.95 ± 0.31 to 698.02 ± 3.38 Bq kg<sup>-1</sup>, 98.68 ± 1.30 to 2397.78 ± 19.64 Bq kg<sup>-1</sup>, and 45.97 ± 2.48 to 2183.80  $\pm$  17.89 Bq kg<sup>-1</sup>, respectively. The absorbed dose rates were above the world average of 59 nGy  $h^{-1}$ . The reported annual effective dose rate outdoor for all the sites ranged from 0.30 to 1.37 mSv y<sup>-1</sup> with average for some areas below 1.00 mSv  $y^{-1}$ ; while other sites had averages higher than unity. They concluded that mining activities led to the increase of the radiation level of the environment, posing a health risk to the inhabitants. A similar result was reported by [4]. In a study by [3] to determine the natural radioactivity levels of primordial radionuclides in soil mine tailings from selected mines in Southwestern Uganda using NaI (Tl) gamma ray spectrometer, it was found that the specific activity concentrations varied from 35.5 to 147.0 Bq kg<sup>-1</sup> for  $^{238}$ U, 119.3 to 376.7 Bq kg<sup>-1</sup> for  $^{232}$ Th and 141.0 to 1658.5 Bq kg<sup>-1</sup> for  $^{40}$ K. The reported mean absorbed dose rates for the studied mines were more than three times the world wide average value of 59 nGy h<sup>-1</sup>. The reported mean outdoor annual effective dose rates for the mines were also more than five times the world average value of 0.07 mSv y<sup>-1</sup>. It was concluded that the mine tailings from the studied areas must not be used as major building material to minimize radiological hazards. A radiometric survey for uranium potential by [8:6] reported extremely high concentrations of uranium in granitic rock outcrop in Gulu and Amuru districts as: 1016.8 Bq kg<sup>-1</sup> for <sup>238</sup>U, 262.4 Bq kg<sup>-1</sup> for <sup>232</sup>Th and 469.5 Bq kg<sup>-1</sup> for <sup>40</sup>K. There was no later analysis to identify the problem of the high background radiation exposure affecting some quarry workers and members of the public and hence remedies were not suggested. A study by Gulu district local government showed that there was limited scientific information from the study areas regarding release of PR into the environment due to the active quarries [9]. Rocks from the quarries were used as building materials without the knowledge of the PR activity concentrations and the potential radiological hazards associated with the building materials [9,11,12]. It was reported that the quarry workers and the local population were not aware of the effects of their quarry works and hence needed to be alerted and encouraged to take precautions [11].

The objective of this study, therefore, was to determine the PR ( $^{238}$ U,  $^{232}$ Th &  $^{40}$ K) activity concentrations, absorbed gamma dose rates (*D*), annual effective dose equivalent outdoor (*E<sub>o</sub>*), excess lifetime cancer risk (*ELCR*), radium equivalent activity (*Ra<sub>eq</sub>*), external hazard index (*H<sub>ex</sub>*), internal hazard index (*H<sub>in</sub>*) and gamma index (*I*) and associated minerals in rock samples collected from selected major active quarries in Gulu and Amuru districts of northern Uganda. This study was done following the report from the district leaders that stone quarry workers needed to be alerted about the effects of the prolonged quarry activities and encouraged to take precautionary measures before individuals were exposed to natural radiation in excess at a threshold level.

### 1.1 Description and Geology of the Study Area

In northern Uganda, the study areas have their local geological conditions dominated by Precambrian Crystalline Basement. It consisted of quartzites, phyllites and undifferentiated acidic gneiss with some intermediate gneiss. The basement rocks were probably of Archean age 2,910 Ma [8, 13]. The major rock types were composed of remnants of low surfaces and scarps related to rift and sediments of the western rift valley. Limited geological investigations were already carried out in some parts of the study areas. In Gulu area, for example, Chalcopyrite copper mineral occurred in granitic gneisses at Lawiyadul area, 10 km north of Gulu City. In Amuru district, there were Kynites in the pennatic vein rock areas of Atiak trading centre while Magnetite (a source of iron ore) were found at Patiri hills in Alero and the surrounding hills. Gold was found in Bibia in Atiak, and through to the border of South Sudan. More minerals were also discovered in rocks of Keyo hills; a few miles north east of Patiri hills [9]. Further geological investigations had shown the presence of quartz, Kfeldspar, plagioclase, hornblende, biotite, and opaque minerals [8]. In many rocks, high radioactivity values were associated with clusters of mafic minerals, for example, biotite in granites or riebeckite in alkalic granites, largely because radioactive accessory minerals were commonly associated with mafic components. Due to the competitive market demands for rock materials by the growing building and construction industries, rock quarries had become a fast income generating activity in the study areas. Most of the miners were widows aged between 22 to 36 years, school dropouts and orphans aged between 10 to 16 years, who were looking for school fees [11, 12]. Granite was the principal lithology extracted although minor amounts of amphibolite were also being exploited [9]. In Gulu, some of the major active quarries during the study were located at Laroo, Lakwana, Kidere, Cwero, Bardege, Palaro. In Amuru district, Palukere, Pacilo, Palema and at Keyo hills, were the active sites [9]. Over 5,000 inhabitants with rudimentary tools depended economically on the quarry works [11].

Unfortunately, majority of them worked without any kind of protective gears [12]. They dug the rocks, broke, and processed them into gravels and aggregates for sale to various constructors, while dust engulfs the air. About 240 tonnes of rudimentarily processed rock ores were reported to be consumed daily from each quarry sites [9].

Gulu District\_headquarters is 332 km by road from Uganda's Capital City, Kampala, with a total land area of 3,499 km<sup>2</sup> (1.44% of the Uganda land size), of which 96.9 km<sup>2</sup> (0.8%) is open water. It is bounded by latitudes  $2^{\circ}10'$  and  $03^{\circ}6'$  N and longitudes  $32^{\circ}10'$  and  $33^{\circ}30'$  E as shown in Figure 1.

The vegetation of Gulu by the time the research works were being done, consisted of intermediate Savannah grassland. This type of vegetation was found between the moist and the dry Savannah. The vegetation type was characterized by open canopy of trees of 10-12 meters high and underlying grasses of 80 centimeters high [9, 18].

The population of Gulu was 436,345 people according to the 2014 Uganda's population census with over 85 % of the people depending on subsistence agriculture [9, 18]. Amuru District headquarters is 400 km by road from Kampala via Gulu, with a total land area of 4851.68 km<sup>2</sup> (2% of the Uganda land size), bounded by latitudes 2°35' and 03°45' N and longitudes 31°30' and 32°10' E as shown in Figure 1 [18]. It is 62 km away from Gulu. Amuru District is endowed with vegetations that covered nearly 24,000 hectares of forests. These include six central forest reserves at Keyo, Olwal, Labala, Got Gweno, Wicheri, and Kilak. There is also a local forest reserve covering three hectares in Pabbo. The district also hosts a 922 km<sup>2</sup> game reserve.

The relief of both Gulu and Amuru for a long time consisted of complex low landscape with relatively uniform topography marked by few sharp contrasts. Oroko and Ajulu hills are in the northern part of Gulu, Ayamo, Awere and Omoro hills are east of Gulu, while Kilak and Keyo hills are found in Amuru District. Generally, the altitude ranges between 1000-1200 m above sea level with minimum temperature of 18 °C and maximum temperature of 35 °C per year [9].

The climate consists of dry and wet seasons, and the average total rainfalls receive is 1,500 mm per annum with monthly average rainfalls varying between 1.4 mm in January and 230 mm in August.

The wet season still extends from April to November with the highest peaks usually witnessed during May, August and October. On the other hand, the dry season begins in November and extends up to March. Relative humidity is high during the wet season and low in the dry season [9].

The population of Amuru was 186,696 people as of 2014 Uganda's population census with over 95 % depending also on subsistence agriculture [9, 18].



Figure 1: Map of Gulu and Amuru districts showing study areas and quarry sites [19].

# 2. Materials and Methods

### 2.1 Sample Collection

The random sampling method recommended by [17] was adopted for equitable representation of the sampling areas. In this method, a quarry site was divided into four (04) sections. Each section was about 30 m apart and from each section four (04) rock samples were collected according to colour and texture using a chisel and later put in a polyethene bag. This same method of [17] was repeated for the remaining four (04) sites until 20 samples were attained. All the samples were then transported to Makerere University, Geology Department, where they were sorted and classified with the help of a senior technician in preparation for analysis.

#### 2.2 Sample Preparation

The samples were oven dried to 120  $^{0}$ C for nine hours until constant weight was attained. This was done to ensure that moisture was completely removed from the sample in order to avoid clamping of the sample particles during crushing [20]. The samples were then crushed using a hammer to obtain fine particles. The particles were then grounded in a mortar using a pestle to powder form in order to increase the surface area and sieved through a 200 µm mesh as stated in [21] to homogenize the contents. Thereafter, the sieved sample was filled in an air tight standard 500 ml plastic Marinelli beaker; labeled with a file name and weighed using a digital meter to determine its (dry) mass. The digital meter registered an average of  $600 \pm 1$  g for each prepared sample. The air tight sealed sample was stored for three weeks to attain secular equilibrium between <sup>226</sup>Ra and decay products of <sup>222</sup>Rn before counting [22, 23, 24].

#### 2.3 Sample Analysis

#### 2.3.1 Detector Calibration

The Sodium Iodide detector (GDM 20 series) was used to detect the gamma rays and their energy as described by [3:6]. The gamma ray detector was first calibrated. The energy and efficiency calibrations of the detector were done using <sup>152</sup>Eu standard source because it emitted many gamma rays of known intensity and energy. A spectrum of <sup>152</sup>Eu was collected for 5000 seconds and saved in the computer hard disc. The peaks of known energies were analyzed using AutoDas software to convert the channel number scale into an energy scale. After the energy and efficiency calibrations, the resolution of the detector was also determined. To obtain the background radiation, a spectrum was generated for a period of 5000 seconds and saved in the computer hard disc. The background spectrum was later subtracted from the gamma ray spectra of the samples to obtain the net counts.

# 2.3.2 Determination of Activity Concentration of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K

Assuming secular equilibrium between <sup>238</sup>U and <sup>232</sup>Th with their progenies, the activity concentrations of <sup>238</sup>U in the samples were computed from the average energies of 351.9 keV of <sup>214</sup>Pb, and 609.3 keV of <sup>214</sup>Bi, respectively. Similarly, the activity concentrations of <sup>232</sup>Th were computed from the average energies of 238.6 keV of <sup>212</sup>Pb, and 583.2 keV of <sup>208</sup>Tl, respectively. The activity of <sup>40</sup>K was determined directly from its 1460.8 keV gamma-ray peaks following the decay of <sup>40</sup>K as described by [25, 26, 27]. Spectra peaks were analyzed one by one using autoDAS command. The cross was placed immediately at the left edge of the peak and *L* was typed to define the cross of the lower mark. The cross of the upper mark was similarly placed at the right edge of the same peak using command *U*. This was followed by typing *CEN* command which gave centroid, standard deviation, *FWHM*, sum between markers and rate. The activity concentration of *i*<sup>th</sup> nuclides from the respective energy peak was computed using the equation:

$$A_i(Bq \ kg^{-1}) = \frac{N_e}{M_i T \varepsilon \rho}, (1)$$

where  $A_i$  = specific activity ( $Bq kg^{-1}$ ) in the *i*<sup>th</sup> sample.  $N_e$  = net peak area of energy , *e*, (Bq).  $M_i$  = dry mass of the *i*<sup>th</sup> sample (kg). T = the sample measurement time (seconds).  $\varepsilon$  =efficiency of the detector (%).  $\rho$  = branching ratio (absolute gamma emmission probability) (%). Equation 1 has been used extensively in the calculation of activity concentrations of PR in Bq kg<sup>-1</sup>, for example, by [20], [28] and [29].

#### 2.5 Determination of Gamma Dose Rates

#### 2.5.1 Absorbed Gamma Dose Rates (D)

The contribution to the external natural radiation exposure of the miners and the inhabitants surrounding the quarry sites by gamma-rays, in the studied rocks was determined using D in the air at 1 m above the ground surface. Uniform distribution of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K (Bq kg<sup>-1</sup>) in the rock samples were assumed in computing, D, based on the guidelines provided by UNSCEAR [17]. It was assumed that the contributions from other naturally occurring radionuclides were insignificant. The computation was done using the formula reported in the study

by [31], as;

$$D(nGy h^{-1}) = 0.462C_U + 0.604C_{Th} + 0.0417C_K, (2)$$

where *D* was the dose rate (nGy h<sup>-1</sup>),  $C_U$ ,  $C_{Th}$ ,  $C_K$  were the activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K, respectively (Bq kg<sup>-1</sup>), and 0.417, 0.604, and 0.462 (nGy h<sup>-1</sup> per Bq kg<sup>-1</sup>) were the dose conversion factors [17].

#### 2.5.2 Annual Effective Dose Equivalent Outdoor (E<sub>o</sub>)

The annual effective dose equivalent for the study area was obtained to show the health effects of those absorbed dose rates on the quarry workers and on the inhabitants surrounding the quarry sites.

In estimating,  $\underline{E}_{\varrho}$ , the absorbed gamma dose rates was converted to  $E_{\varrho}$ , by making use of a conversion coefficient and the outdoor occupancy factor. In the UNSCEAR [17] reports, a value of 0.7 Sv Gy<sup>-1</sup> was used for the conversion of coefficient from absorbed dose in air to the effective dose received by adults and 0.2 for the outdoor occupancy factor. For Gulu and Amuru districts, the inhabitants take about 9 hours outdoor due to the warm climate astride the equator [18]. An outdoor occupancy factor of 0.40 (9.65/24) [3], was used in computing the  $\underline{E}_{\varrho}$  from the following equation as stated by [32, 33]:

$$E_0(mSv yr^{-1}) = D(nGy h^{-1}) \times 8,760h \times 0.375 \times 0.7SvG y^{-1} \times 10^{-6}, (3)$$

# 2.5.3 Excess Lifetime Cancer Risk (ELCR)

The chances of quarry workers and the inhabitants surrounding the quarry sites developing cancer over a lifetime at a given exposure level was determined by *ELCR*. This value predicted the expected number of cancers in the population of the study area due to long term exposure to PR from the quarry works. It was computed using the equation [34]:

$$ELCR = E_0 \times DL \times RF$$
, (4)

where  $E_{\underline{o}}$  is Annual Effective Dose Equivalent (outdoor), *DL* was duration of life (Life expectancy at birth) given as 63.3 years [18], and *RF* was risk factor ( $Sv^{-1}$ ), fatal cancer risk per sievert. For stochastic effects, ICRP 60 used values of 0.05 for the public [34]. The value of *ELCR* was to be less than 0.29 x 10<sup>-3</sup> for radiation hazard to be considered acceptable as reported in the studies by [35, 36].

# 2.6 Determination of Radiological Hazard Indices of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in the Samples

#### 2.6.1 Radium Equivalent Activity (Ra<sub>eq</sub>)

The  $Ra_{eq}$  was used in this study to assess the suitability of the rock samples from the selected quarries as building materials based on the estimation that 370 Bq kg<sup>-1</sup> of <sup>226</sup>Ra, 259 Bq kg<sup>-1</sup> of <sup>232</sup>Th and 4810 Bq kg<sup>-1</sup> of <sup>40</sup>K produced the same gamma dose rate as reported in [3, 15]. The recommended maximum value of <u> $Ra_{eq}$ </u> in all types of rocks should be less than 370 Bq kg<sup>-1</sup> for safer use [37]. This index was computed using the relation stated by [33] as:

$$Ra_{eq}(Bq \ kg^{-1}) = C_U + 1.43C_{Th} + 0.077C_K,$$
 (5)

where  $C_U$ ,  $C_{Th}$ ,  $C_K$  were activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in Bq kg<sup>-1</sup>, respectively.

### 2.6.2 External Hazard Index (H<sub>ex</sub>)

The radiation exposure due to the PR in the rock samples was to be limited to a value that could not cause erythema, skin cancer, and cataracts to the quarry workers and the surrounding inhabitants.

The maximum permissible value of  $H_{ex}$  was limited to 1 mSv yr<sup>-1</sup> and was to correspond to the upper limit of <u>*Ra*eq</u> of 370 Bq kg<sup>-1</sup> [31] for the studied rock samples to be safe for use as building and construction materials. The  $H_{ex}$  was computed using as reported by [1]:

$$H_{ex} = \frac{C_U}{370Bqkg^{-1}} + \frac{C_{Th}}{259Bqkg^{-1}} + \frac{C_K}{4810Bqkg^{-1}} \le 1, (6)$$

where  $C_U$ ,  $C_{Th}$ ,  $C_K$  were activity concentrations of <sup>226</sup>Ra (<sup>238</sup>U), <sup>232</sup>Th and <sup>40</sup>K, in Bq kg<sup>-1</sup>, respectively.

#### 2.6.3 Internal Hazard Index (H<sub>in</sub>)

The  $H_{in}$  was computed to determine whether the use of the rock from the quarries might not cause respiratory diseases such as asthma and cancer due to internally deposited radionuclides. The maximum permissible value of  $H_{in}$  was limited to 1 mSv yr<sup>-1</sup>. It was computed by the equation presented in the report of [32]:

$$H = \frac{C_U}{185Bqkg^{-1}} + \frac{C_{Th}}{259Bqkg^{-1}} + \frac{C_K}{4810Bqkg^{-1}} \le 1, (7)$$

where  $C_U$ ,  $C_{Th}$ ,  $C_K$  were activity concentrations of <sup>226</sup>Ra (<sup>238</sup>U), <sup>232</sup>Th and <sup>40</sup>K, in Bq kg<sup>-1</sup>, respectively.

#### 2.6.3 Gamma Index (I)

This index was used only as a screening tool to check whether the studied rock samples containing PR could meet the dose criterion of  $I \le 0.5$  for materials normally used in bulk such as concrete and the  $I \le 6$  for superficial and other materials with restricted use such as tiles and board. Gamma index was computed using the equation [3]:

$$I = \frac{C_U}{300Bqkg^{-1}} + \frac{C_{Th}}{200Bqkg^{-1}} + \frac{C_K}{3000Bqkg^{-1}}, (8)$$

where  $C_U$ ,  $C_{Th}$ ,  $C_K$  were activity concentrations of <sup>226</sup>Ra (<sup>238</sup>U), <sup>232</sup>Th and <sup>40</sup>K, in Bq kg<sup>-1</sup>, respectively [1, 3,].

#### 3. Results

#### 3.1 Detected Primordial Radionuclides

Table 1 shows the PR detected in the selected rock samples from Gulu and Amuru districts.

Element	Gamma ray	Emitter Radi-	Half Life	Gamma emission	Efficiency of the
	Energy (keV)	onuclides		probability ρ (%)	Detector (%)
<sup>238</sup> U	351.9	<sup>214</sup> Pb	26.8 min.	35.3	5.9
	609.3	<sup>214</sup> Bi	19.9 min.	45.2	9.3
<sup>232</sup> Th	238.6	<sup>212</sup> Pb	10.6 hr.	43.6	13.6
	583.2	<sup>208</sup> Tl	3.1 min.	85.0	3.8
$^{40}$ K	1460.8	$^{40}$ K	1.3 billion years	10.7	2.9

Table 1: Detected PR in the rock samples from the studied areas.

The most significant PR obtained in the analyzed rock samples were radionuclides of the <sup>238</sup>U and <sup>232</sup>Th series, and the radioactive isotope of <sup>40</sup>K. Some other terrestrial radionuclides such as <sup>235</sup>U series, <sup>87</sup>Rb, <sup>138</sup>La, <sup>147</sup>Sm, and <sup>176</sup>Lu that existed at trace levels in all the analyzed samples were ignored. Since man-made volatile radionuclides such as <sup>137</sup>Cs, <sup>90</sup>Sr, and <sup>131</sup>I were not detected in all the analyzed samples, exposures of the quarry workers and the surrounding inhabitants resulting from anthropogenic sources were considered negligible. Hence, the study areas were free from man-made radionuclides originating from atmospheric nuclear testing, underground nuclear testing, nuclear weapons fabrication, nuclear power production, radioisotope production and nuclear accidents at various locations.

# 3.1 Activity Concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in the Five Quarry Sites

The activity concentrations of  $^{238}$ U,  $^{232}$ Th and  $^{40}$ K in each rock sample from Lakwana, Laroo, Kidere, Keyo hills and Palukere quarries were computed using equation (1) and the averages presented in Table 2.

Table 2: Average activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in rock samples from the selected quarries.

Quarry Site	$^{238}$ U ( <i>Bq kg</i> <sup>-1</sup> )	$^{232}$ Th ( <i>Bq kg</i> <sup>-1</sup> )	$^{40}$ K (Bq kg <sup>-1</sup> )
Lakwana	$71.3 \pm 6.3$	138.1 ± 11.5	1155.1 ± 34.8
Laroo	$42.4\pm4.8$	$87.0\pm7.6$	$1435.4 \pm 46.9$
Kidere	$52.9 \pm 4.6$	$87.8 \pm 15.7$	$1102.4 \pm 39.1$
Keyo hills	$50.8 \pm 4.3$	$93.5\pm9.7$	$282.6\pm10.2$
Palukere	$68.2\pm6.9$	$205.0 \pm 11.4$	$595.7 \pm 20.1$
Average	$57.1 \pm 5.4 \; (4.1 - 207.6)$	$122.3 \pm 11.2 (7.0 - 452.0)$	$914.2\pm 30.2(8.6-2055.4)$
World Average	33 (17-60)	45(11-64)	420(140-850)

Figure 2 shows the average activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in the rock samples from the five quar-



Figure 2: Activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in the five quarries.

Table 3: Comparison of the study results with those in similar rock samples from other areas of the world.

Country	Study Area	$^{238}$ U( <i>Bq kg</i> <sup>-1</sup> )	$^{232}$ Th ( <i>Bq kg</i> <sup>-1</sup> )	$^{40}$ K( <i>Bq kg</i> <sup>-1</sup> )	Reference
Kenya	Homa Mountain	195.3	409.5	915.6	[1]
Nigeria	Ibadan Nigeria	12.3	3.8	126.8	[2]
Egypt	Southeastern desert	630.7	550.6	1552.7	[4]
India	Mandya district	30.5	34.4	700.2	[5]
Kenya	Kerio Valley Region	94.5	65.0	1003.9	[6]
Yemen	Shabwah /Hadramout regions	45.0	106.0	1235.0	[7]
Uganda	Amuru, Gulu and Adjumani	1016.8	262.4	469.5	[8]
Egypt	Sol Hamed Area	610.0	110.0	1157.0	[10]
Uganda	Natondome Quarries	51.3	1291.9	1816.9	[15]
Uganda	Kilembe Copper-Cobalt Mines	187.0	19.0		[16]
Czech Republic	Jeronym Mine	161.0	28.0	878.0	[21]
Ghana	Accra Metropolis	12.0	41.0	414.0	[25]
Saudi Arabia	Saudi Arabia	28.8	34.8	665.1	[27]
Palestine	Hebron Governorate	47.6	4.2	100	[29]
World average	-	33.0	45.0	420.0	[17]
Uganda	Gulu and Amuru Quarries	57.1	122.3	914.2	Present study

Table 2 compares the reported average values of PR in similar rock samples analyzed in other areas in Uganda and other parts of the world with the results of this study. The table shows\_that PR in the compared areas varied from one place to another depending on the local geological conditions and each rock characteristics.

#### 3.2 Radiological Hazard Indices

The absorbed gamma dose rate (*D*), annual effective dose equivalent outdoor ( $\underline{E}_0$ ), excess lifetime cancer risk (*ELCR*), radium equivalent activity ( $Ra_{eq}$ ), external hazard indices ( $H_{ex}$ ), internal hazard indices ( $H_{in}$ ), and gamma index (*I*) were computed using equations (2), (3), (4), (5), (6), (7) and (8), respectively. The average values

for each quarry site are presented in Table 4.

Table 4: Average radiological hazard indices in the samples from the selected quarry sites.

Site	$D(nGy h^{-1})$	$E_o(mSv yr^{-1})$	ELCR $x10^{-3}$	$\operatorname{Ra}_{eq}(Bq \ kg^{-1})$	H <sub>ex</sub>	H <sub>in</sub>	Ι
Lakwana	$164.52 \pm 9.94$	$0.40\pm0.03$	$1.27\pm0.09$	$357.73 \pm 18.17$	$0.97\pm0.06$	$1.08\pm0.06$	$1.31\pm0.07$
Laroo	$132.01 \pm 8.75$	$0.32\pm0.03$	$1.02\pm0.08$	$277.38\pm13.29$	$0.75\pm0.04$	$0.86\pm0.05$	$1.06\pm0.05$
Kidere	$104.46 \pm 12.78$	$0.3\pm0.03$	$0.95\pm0.10$	$223.75 \pm 23.52$	$0.61\pm0.08$	$0.72\pm0.07$	$0.84\pm0.09$
Keyo hills	$91.75\pm8.26$	$0.23\pm0.02$	$0.71\pm0.06$	$204.39\pm14.61$	$0.55\pm0.04$	$0.69\pm0.05$	$0.73\pm0.05$
Palukere	$180.14 \pm 10.91$	$0.44\pm0.02$	$1.39\pm0.10$	$407.15 \pm 19.79$	$1.10\pm0.06$	$1.28\pm0.04$	$1.45\pm0.08$
Range	(7.0 - 385.1)	(0.02 - 0.9)	(0.04 - 1.8)	(14.6 - 859.1)	(0.04-2.3)	(0.05-2.9)	(0.06-3.1)
Average	$134.58\pm10.13$	$0.34\pm0.03$	$1.07\pm0.09$	$294.08\pm17.88$	$0.80\pm0.06$	$0.93\pm0.05$	$1.35\pm0.09$
World limit	60.00	0.07	0.29	370.00	1.00	1.00	1.00

#### 3.3 Mineralogical Analysis

The TLM analysis of the rock samples indicated the presence of minerals such as quartz, potassium feldspar, plagioclase, muscovite, biotite, and opaque minerals.

Table 5: The TLM results of associated minerals in the sampled rocks from the selected quarry sites.

Mineral	Volume (%)	Chemical Composition	Description
Quartz	96	SiO <sub>2</sub>	Valid Species Prehistoric.
Potassium Feldspar	35	KAlSi <sub>3</sub> O <sub>8</sub>	Turbid, showed patchy or threadyperthritic tex-
			ture
Plagioclase	20	NaAlSi <sub>3</sub> O <sub>8</sub>	Sodium Plagioclase feldspar. Clear and twined.
Muscovite	5	KAl <sub>2</sub> (AlSi <sub>3</sub> ) O <sub>10</sub> (OH) <sub>2</sub>	Flakes with one perfect cleavage. habit and bire-
			fringence distinctive
Biotite	5	K (Mg, $Fe^{+2}$ ) <sub>3</sub> (Al, $Fe^{+3}$ )	Granitic rocks. Formed a series with phlogopite
		Si <sub>3</sub> O <sub>10</sub> (OH, F) <sub>2</sub>	
Opaque Minerals	10	N/A	Appeared dark to black, subangular, angular, an-
			hedral, crack in most cases, medium to fine grain.

#### 4. Discussion

# 4.1 Activity Concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K

The activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in the rock samples of selected quarry sites in Gulu and Amuru districts are summarized in Table 2. The activity concentration of <sup>238</sup>U ranged from  $4.1 \pm 1.7$  to 207.6 ± 6.5 Bq kg<sup>-1</sup> with an average values of 57.1 ± 5.4 Bq kg<sup>-1</sup>, <sup>232</sup>Th from 7.0 ± 2.0 to 452.0 ± 2.3 Bq kg<sup>-1</sup> with an average values of 122.3 ± 11.2 Bq kg<sup>-1</sup> and <sup>40</sup>K from 8.6 ± 2.0 to 2055.4 ± 73.1 Bq kg<sup>-1</sup> with an average values of 914.2 ± 30.2 Bq kg<sup>-1</sup>. All the average values were higher than the world wide average activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K published by [17], which are 33.0, 45.0 and 420.0 Bq kg<sup>-1</sup>, respectively. This indicates the presence of radioactive minerals in the studied samples. Similar remarkable high values of PR were obtained by other authors [4, 7, 15]. In determining the activity concentrations of <sup>40</sup>K, <sup>232</sup>Th, <sup>226</sup>Ra and radiation exposure levels in the Tabaka soapstone quarries of the Kisii region of Kenya, using a high resolution gamma ray spectrometry, results above the world limits were also reported [28]. They attributed the high values of PR to the present of accessory minerals such as zircon, iron oxides, fluorite and other radioactive related minerals. Such

high activity concentrations of PR in rock samples were reported to be dangerous to the health of miners, constructors and the inhabitants. This is not significantly different from the values obtained by [1, 6]. On the contrary, [2, 25, 29] reported a rather lower values of PR due to sedimentary rock samples. Figure 2 shows comparisons between the average activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in the rock samples collected from the five (05) quarries. The results revealed that the activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K varied from one quarry site to another. The variation of PR in the five quarries was due to variations in rock characteristics and concentrations of PR in the geological formations. Rocks of granitic composition, as pointed out by [8] presented high PR values and low values were presented by sedimentary rocks. The presence of such high PR in studied granitic rocks may be attributed to the presence of quartz, K-feldspar, plagioclase, hornblende, biotite, and opaque minerals. It was also observed that, the mean value of <sup>40</sup>K in Lakwana, Laroo, Kidere, Keyo Hills and Palukere quarries were higher when compared with <sup>238</sup>U and <sup>232</sup>Th. This makes <sup>40</sup>K the dominant PR in all the quarries. In 2016, [3] determined natural radioactivity levels due to mine tailings from selected mines in Southwestern Uganda using NaI (Tl) gamma ray spectrometer. They found out that <sup>40</sup>K occurs in potassium-rich minerals such as feldspars and micas and in most terrestrial materials with an abundance of 0.012%. Our results also show that, of the five (05) quarries, Laroo registered the highest average values of  ${}^{40}$ K (1435.4 ± 46.9 Bq kg<sup>-1</sup>). The major contributor to the high value was granite gneiss rock sample ( $2055.4 \pm 73.1$  Bq kg<sup>-1</sup>). Also, [31] carried out a study on radiological risk associated with naturally occurring radioactive materials (NORM) at selected quarry sites in Abuja FCT, Nigeria, using gamma-ray spectroscopy. The result revealed that granite rocks had high concentrations of  ${}^{40}$ K, which is consistent with the result of our study. Lower average value of  ${}^{40}$ K (282.6 ± 10.2 Bq kg<sup>-1</sup>) was obtained from Keyo Hill. The main contributor to this low value was Novaculate rock sample  $(18.9 \pm 2.8 \text{ Bq kg}^{-1})$ . The highest average concentration of  $205.0 \pm 11.4 \text{ Bq kg}^{-1}$  for <sup>232</sup>Th was found in Palukere and the lowest average value of <sup>232</sup>Th was obtained in Laroo (87.0±7.6 Bq kg<sup>-1</sup>). The main contributors to the high and low values of <sup>232</sup>Th were shale (452.0  $\pm$  2.3 Bg kg<sup>-1</sup>) and Feruginized quartz vein (7.0  $\pm$  2.0 Bg kg<sup>-1</sup>) rock samples, respectively. The average activity concentrations of <sup>232</sup>Th were distinctly higher than values obtained for <sup>238</sup>U in all the quarry sites. This result agrees with previous studies [24, 33]. The <sup>232</sup>Th, according to [23], is about four times more abundant than <sup>238</sup>U in the earth's crust. Lakwana presented the highest average concentrations of  $^{238}$ U (71.3 ± 6.3 Bq kg<sup>-1</sup>), whereas Laroo presented the lowest average value of  $^{238}$ U (42.4 ± 4.8 Bq kg<sup>-1</sup>). The high average value of  $^{238}$ U in Lakwana was due to calcarenite rock sample (207.6 ± 6.5 Bq kg<sup>-1</sup>) <sup>1</sup>) and the low average value of <sup>238</sup>U in Laroo was due to gneiss rock sample (15.5  $\pm$  5.6 Bq kg<sup>-1</sup>). The average activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in each of the five (05) quarries were all higher than the world wide average of 33.0 for <sup>238</sup>U, 45.0 for <sup>232</sup>Th, and 420 Bq kg<sup>-1</sup> for <sup>40</sup>K reported by [17]. This shows that the quarry works pose a radiological hazard to the miners, and local inhabitants. Also, the results obtained in this work were compared with studies by other authors in Uganda and other parts of the world for similar rock types as presented in Table 3. This was done to ascertain whether there were significant variations in the activity concentration of PR with the different geological conditions. In a study of geology and geodynamic development of Uganda, [8] reported higher average value 1016.8 Bq kg<sup>-1</sup> for <sup>238</sup>U, which was 17.8 times higher than the results of this study. The study argued that the uranium anomaly was caused by a pegmatite intrusion within the sheared topographically high Adjumani granite trending NW-SE. In another developmental study, [15] reported the maximum values 1291.9 Bq kg<sup>-1</sup> of <sup>232</sup>Th, and 1816.9 Bq kg<sup>-1</sup> of <sup>40</sup>K in Natondome quarries in Mbale District in Eastern Uganda. The reported values were greater than the present work by a factor of 10.6 and 2, respectively. They attributed the high values to the quarry being a high background radiation area. Also, [2] determined the activity concentrations in selected rock samples from quarry sites in Ibadan, Nigeria. Analysis of their measurements showed lower values 12.3, 3.8, and 126.8 Bq kg<sup>-1</sup> of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K, respectively. These values were 4.6, 32.2, and 7.2 times lower than the average values obtained in this study but compare well with values obtained for Novaculite rock in Keyo Hills quarry. Analysis of Table 3 shows that the PR varied from one area to another and from one country to another as determined by their local geological conditions.

#### 4.2 Radiological Hazard Indices

The average values of the radiological hazard indices in the samples from the selected quarry sites are presented in Table 4. From Table 4, The results of the computed absorbed gamma dose rate (D) in all the rock samples varied from 7.0 (Feruginized quartz vein from Keyo hills) to 385.1 nGy h<sup>-1</sup> (calcarenite rock from Lakwana) with an average value of 134.6 nGy  $h^{-1}$  which is 2.2 times higher than the world wide average of 60 nGy  $h^{-1}$ [31]. This was a pointer to the fact that the quarry workers and the inhabitants were significantly exposed to gamma rays from the detected PR in the quarried rocks. This is also in agreement with [10] who reported higher absorbed dose rate (D) in the air of 396 nGy  $h^{-1}$  being 6.6 times above the acceptable world limits. They acknowledged that it was not safe to use the studied rocks in building and infrastructure applications. The corresponding annual effective dose equivalent outdoor  $(E_o)$  varied from 0.02 (Novaculite, dolomite, quartz sandstone) to 0.9 mSv yr<sup>-1</sup> (calcarenite rock from Lakwana) with an average of  $0.3 \pm 0.1$  mSv yr<sup>-1</sup>. This average value was 4.3 times higher than world limit of 0.07 mSv yr<sup>-1</sup> for  $E_0$  [32]. As shown in Table 4, average values of  $E_0$ in all the studied quarry sites were beyond the world limit of 0.07 mSv yr<sup>-1</sup> reported by [30]. The highest average value of  $E_0$  was found in Palukere quarry and the lowest found in Keyo Hills quarry. The acceptable  $E_0$  recommended for members of the public without constraint is 1.0 mSv yr<sup>-1</sup> for safety purposes [17]. The results for  $E_{0}$ , is indicative of a significant health effects of D, on the quarry workers and the inhabitants. Also, [29] in the study of radionuclides measurements in some rock samples collected from the environment of Hebron Governorate –Palestine pointed out that the values of  $E_0$  varied from 0.0138 to 0.0635 mSv y<sup>-1</sup> with average value of 0.0336 mSv y<sup>-1</sup>. They further asserted that the obtained values of  $E_0$  from the study were all lower than the accepted average worldwide value of 0.07 mSv yr<sup>-1</sup>. In this study, the radium equivalent activity ( $Ra_{eq}$ ) were also evaluated. The  $Ra_{eq}$  is a single quantity that compares the activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in rock samples in order to obtain a total activity concentration. Table 4 shows that the average value of  $Ra_{eq}$  ranged from 14.6 to 859.1 Bq kg<sup>-1</sup> with an average of 294.1  $\pm$  17.9 Bq kg<sup>-1</sup>. According to Table 4, the values of  $Ra_{ea}$  for all the quarry sites were much less than maximum limit of 370 Bq kg<sup>-1</sup> as recommended by the Organization for Economic Cooperation and Development [37], except that of Palukere quarry which is above the accepted safe limit. Hence, the rocks from the studied areas were safer for use as building materials except rocks from Palukere quarries. Also, as indicated in Table 4, the computed values of the external hazard index  $(H_{ex})$  for all the quarry sites varied from 0.04 to 2.3 with an average value of  $0.8 \pm 0.1$ . The values of  $H_{ex}$  for all the quarry sites except Palukere were below unity ( $H_{ex} < 1$ ). This meant long term use of rocks (normally above 10 years) as building materials from all other quarries were safer than rocks from Palukere quarries that could cause workers and inhabitants diseases such as cancer of the skin, lungs, female breast, bones, and thyroid [3, 15, 36]. In contrast, [30] determined radioactivity levels and hazards of sediment and rock samples in İkizdere and Kaptanpaşa Valley, Turkey. Their report showed that the computed values of  $H_{ex}$  ranged from 0.20 to 0.32 with

an average of 0.27. They concluded that the health effect due to natural radiation was insignificant. Borrowing a leaf from Table 4, the computed values of internal hazard index  $(H_{in})$  for all the quarry sites were in the range of 0.05 to 2.9 with an average of  $0.9 \pm 0.1$ . The average value of  $H_{in}$  for all the samples from Kidere, Laroo, Keyo hills were well below the safe limit of 1 set by [17]. However, the values of  $H_{in}$  from Palukere and Lakwana were above unity  $(H_{in} > 1)$ . Much as the rocks from Kidere, Laroo, and Keyo hills quarries were relatively safer from posing radiological hazard health risks, long term use of rocks from Palukere and Lakwana could still pose such health risks. Also, the computed values of gamma index (1) as shown in Table 4 varied from 0.06 to 3.1 with an average value of 1.4  $\pm$  0.1. The value of *I* for Lakwana, Laroo, and Palukere quarries were all above  $I \leq$ 1 but within the acceptable limit of  $I \le 2$  for superficial and other materials with restricted use [3:158, 33:41]. Using recommendations from [37], rocks from Lakwana, Laroo, and Palukere quarries fell within materials that should not be used in bulk but rather moderately to prevent gamma radiation exposure hazards. Although values of I for Kidere and Keyo hills quarries were within the acceptable limit of  $I \le 1$  for materials used in bulk, any dose, no matter how small, would produce some risks. Radiation exposures by workers and the inhabitants in the study areas were to be held to the absolute minimum (ALARA working principles). As seen in Table 4, the range of ELCR for the studied quarry sites was from 0.04 x 10<sup>-3</sup> to 1.8 x 10<sup>-3</sup>, with an average of 1.0 x 10<sup>-3</sup>. This average value was 3.4 times higher than the world average of  $0.29 \times 10^{-3}$  set by [17], supported by later studies conducted by [3, 33, 36]. This meant that the radiation exposure had the potential of causing hereditary effects in the off-springs of 310 persons from Gulu and 133 persons from Amuru districts, respectively. This kind of exposure therefore could lead to cancer of the breast, prostate cancer, or even blood cancer [38, 39]. And [36] equally carried out PR concentration and ELCR due to gamma radioactivity from quarry stone aggregates in Jos and its surroundings, North Central Nigeria. They reported *ELCR* value ranging from  $3.21 \times 10^{-3}$  to  $5.68 \times 10^{-3}$ , with an average of  $4.45 \times 10^{-3}$ . The reported average value was 15.34 times higher than the world average of  $0.29 \times 10^{-3}$  set by [17]. The report concluded that crushed stones from quarries in Jos and its surroundings might have contributed significantly to cancer risks in the area.

#### 4.3 Associated Minerals

Quartz (SiO<sub>2</sub>), potassium feldspar (KAlSi<sub>3</sub>O<sub>8</sub>) and opaque minerals were the main minerals found in most of the samples as shown in Table 5. Similar results were reported in other studies [40, 41]. The SiO<sub>2</sub> appeared in all the samples because it was found to be the commonest mineral on the earth's surface and highly resistant to chemical weathering [40, 41 - 45]. This possibility was studied in more details by [8, 40, 41]. The SiO<sub>2</sub> when accumulated and consolidated as deposits of sand form sandstones, an important group of sedimentary rocks. The KAlSi<sub>3</sub>O<sub>8</sub> was not detected in ferruginized quartz vein.

The KAlSi<sub>3</sub>O<sub>8</sub> was found to be relatively and easily decomposed by weathering. The alkali element was easily carried away in solutions. Other minerals detected were plagioclase (NaAlSi<sub>3</sub>O<sub>8</sub>), muscovite (KAl<sub>3</sub>Si<sub>3</sub>O<sub>10</sub> (OH)<sub>2</sub>), biotite (K(Mg,Fe)<sub>3</sub>AlSi<sub>3</sub>O<sub>10</sub>(OH)<sub>2</sub>), and iron oxides (FeO<sub>3</sub>) which were accessories. These results were similar with those from [8, 40], but slightly different from the results from [42, 43 - 45]. Our data showed that high concentrations of PR were in samples with low amount of SiO<sub>2</sub> but had high amount of KAlSi<sub>3</sub>O<sub>8</sub> and opaque minerals. The low concentrations of PR in the samples with high amount of SiO<sub>2</sub> on the other side had reduced amount of opaque minerals.

#### 5. Conclusions

This study was one of the most recent researches carried out in the field of radiation physics in Uganda. Its major focus was to assess the radiological hazards and the associated risks on quarry workers, users of building materials, and on the surrounding inhabitants in the two districts of Gulu and Amuru. The findings were scientifically done using Na (Tl) Gamma ray Spectrometer and Transmitted Light Microscope. The average activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K were all above the world limit. These indicate the presence of radioactive minerals in the studied samples. The activity concentrations of the PR varied from one quarry site to another. The variation of the PR in the five quarries was due to variations in rock characteristics and concentrations of the PR in the geological formations. Rock samples of granitic composition presented high PR values and low values were presented by sedimentary rocks. The presence of such high PR in the studied samples are attributed to the detected minerals. Mineralogical analyses show the presence of quartz, potassium feldspar, plagioclase, biotite, muscovite, and opaque minerals. Quartz was the major mineral while the rest were accessories. The mean values of <sup>40</sup>K in the five quarries were higher when compared to <sup>238</sup>U and <sup>232</sup>Th. Hence, <sup>40</sup>K was the dominant PR in all the quarries. Also, the average values of D,  $E_o$  and ELCR for the five quarries were above the world limits. However, average values of  $Ra_{eq}$ ,  $H_{ex}$ , and  $H_{in}$  were below the world limit except for Palukere quarry. The study has established that radiological hazards from the quarries posed high health risks to quarry workers, users of the rocks, and the surrounding inhabitants. Also, prolonged radiation exposure had the potential of causing cancer related diseases to the off-springs of 310 persons from Gulu and 133 persons from Amuru district. Therefore, the study recommended that quarry works be regulated to avoid infections resulting from persistent manual work at the quarry sites.

#### 6. Recommendations

The results of this study could be used by different institutions such as Uganda Atomic Energy Council (AEC) that protects the environment from dangers resulting from ionizing radiation. This body could assist to advise quarry workers on good practices. The National Environmental Management Authority (NEMA) that controls environmental systems to ensure that Gulu and Amuru should work in line with set principles on environment and where necessary to enforce the quarry by-laws and ordinances. To urge the Directorate of Geological Survey and Mines (DGSM) to also conduct geological studies for valuable radioactive minerals in the study areas of Gulu and Amuru district. The Uganda Ministry of Health (MoH) experts could also carry out additional studies to establish the correlation of cancer infections and quarry works. This would inform the government on corrective measures that protects the population of the region.

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#### 7. Conflict of Interest

The authors declare that they have no conflict of interest.

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